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(54) Title: PLASTIC-BASED MATERIAL			
(57) Abstract <p>A plastic-based sheet-like material (1) contains in a blend a plastic matrix, which contains a chlorine-free polyolefin or mixture of polyolefins, and a plasticizer. The plasticizer of the polyolefins or mixture of polyolefins is an oligomeric poly-α-olefin type substance. The material forms the covering of a structure, especially a floor covering.</p>			

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Plastic-based material

The invention relates to a plastic-based material according to the preamble of the appended claim 1.

5

Polyolefin-based plastic products form a large group with various uses especially as parts of different structures. In this context, slightly deviating from normal practice and for reasons of brevity, polyolefins are used to refer to all such thermoplastic plastics, in which the carbon skeleton of the polymer is formed by polymerizing monomers containing double bonds between carbons, wherein said double bonds open in the polymerization to form carbon-carbon bonds between the monomers. Polyolefins include, according to this definition, for instance, polyethylene, polypropylene, ethylene-propylene copolymer rubber (EPR), styrene-butadiene rubber (SBR), ethylene-butylacrylate copolymer (EBA), ethylene-methylacrylate copolymer (EMA), and ethylene-vinylacetate copolymer (EVA).

20 In several products these polyolefins form a matrix which binds the product into a solid structure. By means of the thermoplastic matrix it is also easy to fabricate the product into a desired shape with processing methods known in the plastic technology, based on the processing of melt plastic material.

25 Matrix plastic forms the framework for the product, and the mechanical properties of the product are primarily dependent thereon. The properties of one plastic grade are rarely suitable, and therefore the aim is to mix compatible polymers in order to achieve optimal properties. In addition, mere polymers or polymer mixtures as such 30 do not bring about the desired properties, but in different products several additives are used, of which extenders, i.e. plasticizers, form an important group. If flexibility and "pliancy" is required of the end product, the plasticizers are very important. The plasticizers should, however, be such substances which can be mixed with the matrix in 35 large quantities for creating the desired effect, without them starting to leak out of the matrix in the course of time.

Flexibility and pliancy are required of products used as parts of structures, especially of coatings, such as floor coverings, which can be laid on different supports. There are several properties required of a floor covering, for the simultaneous implementation of which it is

5 difficult to find a suitable matrix plastic. First, the material has to be easily processable into a sheet-like product, especially extrudable, calenderable, easily pressable to shape or also injection mouldable. It also has to be wear-resistant. Furthermore, its surface properties have to be suitable, i.e. it has to be soil repellent and/or easily coated with

10 different protective coatings. If the sheet-like product is in the shape of a mat which can be rolled up, it has to lay down easily on the support and remain in the deflected shape without a strong tendency to revert into its original shape. Important properties with respect to the use of the product include elasticity in thickness direction (the reversibility of

15 the surface after a point-like load) and a certain softness considering walking comfort. Earlier, polyvinyl chloride (PVC) has been used for fabricating floor coverings, but nowadays the aim is to replace PVC with chlorine-free polymers, such as chlorine-free polyolefins.

20 Finnish patent 94438 presents a plate or mat shaped floor covering, the matrix of which is ethylene-butylacrylate copolymer (EBA) which can be mixed with different propylene polymers and copolymers. The product does not contain liquid plasticizers.

25 Swedish published examined application 503631 also presents a floor covering replacing PVC, the matrix of which contains ethylene- α -olefin copolymer, polypropylene, and cross-linked ethylene polymer. Plasticizers are not mentioned here either.

30 European patent publication 46536 presents a composition which is primarily suitable for the insulating bottom layers of car floor mats and contains ethylene- α -olefin copolymer as a matrix, possibly mixed with ethylene-propylene-diene monomer rubber (EPDM) for increasing extensibility, mineral filler, and plasticizer. As suitable plasticizers the

35 publication mentions paraffin oils, aromatic oils and naphthene oils, under the common name processing oils, epoxidized oils, such as epoxidized soya bean oil and linseed oil, liquid polyesters and liquid

polyethers and polyether esters. The amount of the plasticizers in the total weight of the composition varies from 2 to 12 wt%.

In addition, polyolefin-based matrix plastic mixtures are presented in 5 European patent 428153, which presents, for example, a mixture suitable for the manufacture of sheet-like products and which contains propylene polymer material, amorphous ethylene-propylene copolymer rubber (EPR), semi-crystalline, substantially linear ethylene-propylene copolymer with an ethylene proportion of over 90%, and a vulcanization 10 system. It is possible to use paraffin and naphthene oils as an extender or plasticizer for the thermoplastic elastomer consisting of these substances. European patent 479171, for its part, presents a plastic matrix suitable for manufacturing discrete injection-mouldable products and extruded profiles, which is a mixture of graft copolymers of 15 propylene polymer and graft copolymers of olefin rubbers (EPR), which can also contain conventional propylene polymer mixed with it. This publication does not discuss plasticizers.

The purpose of the invention is to produce a plastic-based material, 20 having mechanical properties suitable for the planar coverings of structures, being based on a chlorine-free polyolefin or mixture of chlorine-free polyolefins and having the properties which can be more freely adjusted by means of a plasticizer. Especially due to properties achieved with specified mixtures of substances, it is particularly suitable 25 to be used as a wide, sheet-like covering for structures, especially as a floor covering.

For achieving the aforementioned purposes, the material according to 30 the invention is primarily characterized in what will be presented in the characterizing part of the appended claim 1.

As a plasticizer for the polyolefin in the matrix of the covering material, 35 an oligomeric poly- α -olefin type substance was found. The properties of the substance are advantageous in connection with several polyolefins and mixtures of polyolefins. It can be blended in large quantities in the matrix without the substance leaking out, wherein the mechanical properties of the substance can be adjusted more freely. This is particularly important if the increase in the flexibility of a sheet-like

product with constant thickness is desired, wherein the plasticizer content is relatively high. The poly- α -olefin type substance is, however, useful also in low contents.

- 5 The invention is especially well suited for materials in which the plastic matrix contains a thermoplastic polyolefin elastomer, such as ethylene- α -olefin copolymer rubber. This polyolefin rubber can be mixed with a non-elastomeric polyolefin homopolymer or copolymer, such as polypropylene. In addition, it is possible to achieve an advantageous
- 10 synergistic effect using a filler and/or reinforcement together with the plasticizer.

In the following, the invention will be described in more detail with reference to the appended drawing which shows a material according to the invention in a cross-section.

The material marked with the reference number 1 in the drawing is a sheet-like covering with constant thickness, which can be attached on a structure 2 with a suitable fastening method. The coating can be composed of plates (tiles) laid adjacent to each other or of a mat-like product spread on the structure from a roll.

In the following, all the feasible components of the material will be described.

25

Matrix

The main component of the plastic matrix is a chlorine-free polyolefin. All thermoplastic polyolefins which contain a long-chained, continuous carbon skeleton produced from double bond monomers by polymerization are feasible. The main component can also be a mixture of such polyolefins. The melting and softening temperatures of the polymers are typically above 70°C and they are processable in melt form. Such chlorine-free polyolefins include: 1) Different polyethylene grades and ethylene copolymers, such as those containing a small amount (less than 10 wt%) of butene or propylene comonomer; propylene-based polymers, i.e. different propylene homopolymers and random copolymers, the latter containing ethylene or C₄-C₁₀- α -olefin

as a comonomer in a small content, usually less than 20 wt%; polybutene-1, 2) Thermoplastic polyolefin elastomers, i.e. polyolefin rubbers, such as ethylene- α -olefin copolymer rubber, for example copolymer rubber containing e.g. ethylene and propylene (EPR), 5 including a) a two monomer copolymer (EPM) and b) a terpolymer (EPDM) containing a diene monomer in addition to these two. The ethylene-butylene copolymer rubber (EBR) and the ethylene-propylene-butene-terpolymer rubber (EPBR) are also ethylene- α -olefin rubbers. According to the definition used here, polyolefin rubbers also include 10 propylene-butene copolymer rubber (PBR) as well as polymers where the side chains contain aromatic parts, such as styrene-butadiene rubber (SBR).

15 Chlorine-free polyolefins include, in this context, also polymers where the side chains contain ester bonds, such as EBA, EMA, and EVA.

In a particularly advantageous manner, the plastic matrix contains thermoplastic polyolefin elastomer, such as the aforementioned polyolefin rubbers. In such compositions, the thermoplastic elastomer is 20 advantageously in a mixture together with a non-elastomeric polyolefin, typically with one of the aforementioned polypropylenes. The mixture can contain in the non-elastomeric component two or more polyolefins, such as propylene-based homopolymer and/or copolymer together with any of the aforementioned polyolefins. Similarly, an elastomer 25 component can contain two or more polyolefin rubbers. Blending propylene-based polymers and polyolefin elastomers together is commonly used to achieve optimal material properties, and in this context in addition to the aforementioned publications reference can be made to the U.S. Patents 5552482, 5360868 and 5143978. In this 30 purpose, the thermoplastic elastomers also include plastomers, the blending of which into polypropylene is described for example in the article "Modifying Polypropylene with a Metallocene Plastomer" by Dharmarajan and Yu in Plastics Engineering, Aug. 1996, pp. 33-35.

35 The elastomer component can also contain cross-linking, and the non-elastomeric component and/or the elastomeric component can also be a graft polymer. Also other components of the material, such as

polyethylene, can be cross-linked. Cross-linking can be conducted by means of a peroxide or other cross-linking agents of prior art.

In addition to one or more polyolefins, the plastic matrix can contain

5 another polymer, for example polyamide, blended together with polyolefin. In such mixtures it is possible to use a compatibilizer for making the non-polyolefin polymer compatible with the polyolefin polymer.

10 All the aforementioned blend alternatives are processable in melt form for producing sheet-like coating materials.

The thermoplastic elastomer includes, in this context, all the polymers which, due to their polymer structure, can be used as structural

15 materials for thermoplastic elastomers, either on their own or as a mixture together with another polymer, and it is possible that they are cross-linked in order to influence the elastomer properties.

Plasticizer

20 The new plasticizer used in the material for the polyolefin of the matrix plastic is a poly- α -olefin type oligomer, the methods of preparation of which are described, for instance, in European application publication EP-442656, in U.S. patent 4,032,591 and in European patent EP-

25 318186. These publications present the use of the substances as lubricants and hydraulic oils. Poly- α -olefin type oligomers are produced by oligomerizing α -alkene monomers, wherein the result is a substance which is liquid (not wax-like) at room temperature, and where a carbon chain, i.e. the core of the oligomer, is composed of carbons

30 connected to each other after the double bonds have opened. In the oligomerization process, suitable catalysts are used, after which the reaction product is still subjected to hydrogenation in order to remove the remaining double bonds. The methods of preparation belong to prior art, and they will therefore not be discussed in detail here.

35 The monomers used contain at least three carbon atoms, preferably at least six carbon atoms, such as 1-decene, mentioned in the U.S. patent 4,032,591 and in the EP patent 318186. The most suitable

number of monomer carbon atoms is from six to twelve. Poly- α -olefin type oligomer has a strongly branched carbon chain. This fact may have significance for its suitability as a plasticizer for the plastic matrix polyolefin. Depending on the degree of oligomerization, the greatest 5 part of the oligomer is a trimer, tetramer, pentamer, hexamer, or a mixture of these. Especially, the content of the tetramer, pentamer, hexamer, or a mixture of them is typically above 75 wt%. Thus, the substance in question is not a polymer, but an oligomer whose chain length is precisely defined and restricted.

10

Other substances

In addition to the plastic matrix and plasticizer, the plastic-based material can also contain a filler and/or reinforcement. As a filler it is 15 possible to use many powdery mineral fillers of prior art, such as calcium carbonate, such as comminuted limestone, e.g. dolomite powder, aluminium trihydroxide, or a mixture of calcium carbonate and aluminium trihydroxide, wherein also good flame proofing is achieved. Other possible fillers include chalk, kaolin, different silicate mineral 20 powders and quartz powders or quartz sand. Fillers can constitute 20-95 % of the total weight of the material.

It is also possible to use fibrous fillers and/or reinforcements known in the reinforced plastic technology. Especially particulate mineral fillers 25 are suitable for covering materials of structures, such as floor coverings, which would be stiffened too much if fibrous fillers were used.

The branching of the poly- α -olefin type oligomer molecules may be 30 important also if the material contains a filler, because it can be assumed that the oligomeric poly- α -olefin partly bonds also to the filler particles, which improves the internal cohesion of the product and the plasticizer will be better retained in the material as well.

35 Naturally, a plastic-based material can contain a small amount of other additives known in the plastic technology, such as antistatic substances, light stabilizers, antioxidants and colouring agents. The

content of these additives, usually used in small quantities, is typically max. 5 wt% of the total weight of the material.

When the covering material of a structure is produced of the

5 aforementioned components, they are blended with each other and the product is made in the manufacture by methods known as such. When sheet-like coverings for the structures are manufactured, the components are mixed, the mixture is heated in order to melt the plastic matrix, after which the mixture is shaped by extrusion or a mixing mill

10 into a wide-surface sheet, which is passed through a two-roll mill and is calendered or pressed in order to produce the final sheet-like product, which can be processed further (cutting, compressing) in order to produce plates. By means of extrusion, it is possible to form the material also into flexible, mat-like products which can be rolled up.

15

On the surface of the coating, there can also be a thin, protective coating layer which is advantageously transparent, because the body typically already provides the product with the desired appearance. In the drawing, this thinner coating layer is marked with the reference number 1b, and the thicker body layer underneath the same, which contains the aforementioned components characteristic to the invention, is marked with the reference number 1b. In floor coverings, this coating layer is primarily intended to prevent scratching.

20

25 The proportion of the plasticizer with respect to plastic matrix can vary greatly depending on the use of the material. The proportion of the oligomeric poly- α -olefin in weight parts per 100 weight parts of the matrix plastic is typically not higher than 160, preferably between 0.5 and 100. At the upper limit 160, the plasticizer may already leak out of

30 the product, but that is not, depending on the application, necessarily disadvantageous. The lower limit of the plasticizer is 0.1 weight parts.

In the following, we shall describe tests conducted with certain materials.

35

In the tests, plasticizers of prior art, recommended for polyolefins, and new, oligomeric poly- α -olefin type plasticizers were used with different plastic matrices.

As oligomeric poly- α -olefin type plasticizers Nexbase 2004 and 2008 oils by Neste Oy were used, which are based on decene monomers. Nexbase 2004 and 2008 differ from each other primarily with respect to the viscosity index, which is typically 123 for the former and 137 for the latter. The kinematic viscosity of Nexbase 2004 is from 3.8 to 4.1 cSt at 100°C, and at 40°C it is from 17.0 to 18.8, whereas with Nexbase 2008 the corresponding values are 7.7 - 8.2 and 46.1 - 50.5. The substances are clear, oily liquids, with a density from 810 to 840 kg/m³ at 15°C. Considering several plastic product applications, it is important that the substances in question are non-toxic and do not irritate the skin and the eyes.

15 The sheets of different matrix plastics were produced in the following
way:

Matrix	1: EBA, LOTRYL 17 BG 04, acrylate content 15 - 18 % (Elf Atochem)
"	2: EEA, OE 5810, acrylate content 16 % (Borealis)
"	3: EMA, OPTENE 5614, acrylate content 14 % (Borealis)
20	4: POE, 8501, ethylene-octene copolymer (Dow Chemicals)
"	5: ADFLEX Q 100F, a mixture of polypropylene and ethylene- α -olefin copolymer (Montell)
"	6: Polypropylene, RB 501 F (Borealis)

The following blending was conducted for the aforementioned matrix plastics:

Blend	C: No filler, no plasticizer
-"-	D: No filler, poly- α -olefin as a plasticizer (Nexbase 2008)
30	7.5 wt% of the total weight of the material
-"-	E: No filler, "Citroflex" as a plasticizer 7.5 wt%
-"-	F: Comminuted limestone as a filler (Parfill) 60 wt%, no plasticizer
-"-	G: Comminuted limestone as a filler (Parfill) 60 wt%, poly- α -olefin as a plasticizer (Nexbase 2008)
35	H: Comminuted limestone as filler (Parfill) 60 wt%, "Citroflex" as a plasticizer 7.5 wt%

Flexural test:

The samples were subjected to a flexural test in the following way: A piece cut off from the sample material is attached at its other end so that a 25 cm long and 3 cm wide strip of the piece is left free, and it is allowed to bend down freely due to the effect of gravity from the horizontal position downwards. The ideal is an even deflection downwards relatively fast into a wide angle. The following table presents the angles with respect to the vertical plane at different moments of time (in the period of 0 s, the angle = 0°). In the table, the combination of a number and a letter indicates unambiguously the matrix plastic and the blend used in it.

5

10

15

The results show that with a poly- α -olefin plasticizer it is possible to achieve ideal properties with the matrix plastic 5 in mat-like floor coverings which can be rolled up.

Table 1. Flexural test results for different samples. The dash indicates a sample, in which the plasticizer has exuded as beads on the surface of the sample material.

Sample	Flexural angle in degrees						
	5 s	10 s	30 s	60 s	120 s	180 s	240 s
C1	36	37	38	38	39	39	39
C2	41	42	43	43	44	45	45
C3	38	39	39	40	41	41	41
C4	66	66	67	67	67	68	68
C5	15	16	17	18	20	20	20
C6	4	4	4	5	5	5	5
D1	44	45	46	46	46	47	47
D2	49	49	50	50	51	51	51
D3	49	50	51	51	51	52	52
D4	70	70	70	71	71	72	72
D5	30	32	33	35	37	37	38
D6	3	3	3	3	4	4	4
E1	-	-	-	-	-	-	-
E2	41	42	43	43	43	44	44
E3	-	-	-	-	-	-	-
E4	-	-	-	-	-	-	-
E5	12	13	14	15	16	17	17
E6	1	1	1	1	1	1	2
F1	11	11	12	12	13	13	13
F2	15	15	15	16	16	16	16
F3	10	10	10	11	11	11	12
F4	36	37	37	38	39	40	40
F5	5	5	6	6	7	7	8
F6	5	5	5	5	5	5	5
G1—G3	-	-	-	-	-	-	-
G4	55	56	57	57	58	59	60
G5	26	27	29	32	33	35	35
G6	5	6	6	7	7	7	8
H1—H5	-	-	-	-	-	-	-
H6	6	6	7	7	8	8	8

The samples were subjected to an accelerated ageing at 60°C and examined at the following points of time counted from the beginning of the test: day 0, day 2, week 1, week 2, week 4, month 1, month 2, month 4, month 8.

5

The surface tension was measured according to ASTM D2578-67 from one day and one week old samples, and the results are presented in Table 2.

10 Table 2. The surface tension of the samples (dynes/cm). e = exuding (plasticizer exuded as beads on the surface of the sample)

Sample	Surface tension		Sample	Surface tension	
	1 day	1 week		1 day	1 week
C1	36	36	F1	38	39
C2	37	38	F2	37	38
C3	38	38	F3	36	35
C4	34	34	F4	33	33
C5	33	33	F5	32	34
C6	32	33	F6	35	34
D1	33	e	G1	e	e
D2	34	e	G2	e	e
D3	34	e	G3	e	e
D4	34	34	G4	33	34
D5	36	34	G5	33	33
D6	35	34	G6	34	34
E1	e	e	H1	e	e
E2	40	e	H2	e	e
E3	e	e	H3	e	e
E4	e	e	H4	e	e
E5	39	e	H5	e	e
E6	35	34	H6	34	34

15 The following table presents test conducted with a particular matrix plastic blend. The matrix Hifax CA12A contains a blend of polypropylene and ethylene- α -olefin copolymer rubber, and the EBA is ethylene-butylacrylate copolymer.

Table 3. Properties of the samples containing filler and matrix plastic with different plasticizer contents.

5

Blends	1 wt%	2 wt%	3 wt%
Hifax CA12A (Montell)	19.8	19.4	18.8
EBA 6417 (Borealis)	11.9	11.6	11.3
Nexbase 2004 (Neste)	0	1.9	4.7
Parfill S9 (limestone)	59.3	58.1	56.5
ATH, Baco FRF10	7.1	7	6.8
TiO ₂	1.9	2	1.9
Tensile strength/MPa/	2.4	2.5	2.8
Strain %/	11.6	13.5	19.3
Tensile modulus /MPa/ (Young)	450	366	354

10

The table shows that the plasticizer used has an advantageous effect on the strength properties of the samples, i.e. the strength and strain grow and the modulus decreases when the plasticizer content is increased.

Claims:

1. A plastic-based, sheet-like covering material (1) for a structure, especially a floor covering, which contains in a blend
 - 5 - a plastic matrix comprising a chlorine-free polyolefin or mixture of polyolefins, and
 - a plasticizer,

characterized in that the plasticizer of the polyolefin or mixture of polyolefins is an oligomeric poly- α -olefin type substance.

10 2. A covering material according to claim 1, characterized in that it is a mat-like covering material which can be rolled up.

15 3. A material according to claim 1 or 2, characterized in that the material contains a filler and/or reinforcement, for example a powdery mineral filler.

20 4. A material according to any of the claims 1 to 3, characterized in that the poly- α -olefin molecules are composed of monomers of at least three carbon atoms, preferably of monomers of at least six carbon atoms.

25 5. A material according to claim 4, characterized in that the poly- α -olefin molecules are composed of monomers of six to twelve carbon atoms.

30 6. A material according to any of the previous claims, characterized in that most of the poly- α -olefins is a trimer, tetramer, pentamer, hexamer, or a mixture thereof.

35 7. A material according to any of the previous claims, characterized in that the content of the oligomeric poly- α -olefin is 0.1 - 160 weight parts, preferably 0.5 - 100 weight parts per 100 weight parts of the plastic matrix.

8. A material according to any of the previous claims, characterized in that the plastic matrix contains a thermoplastic polyolefin elastomer, such as ethylene- α -olefin copolymer rubber.

9. A material according to claim 8, **characterized** in that the thermoplastic polyolefin elastomer is blended with a non-elastomeric polyolefin homopolymer or copolymer, such as polypropylene.

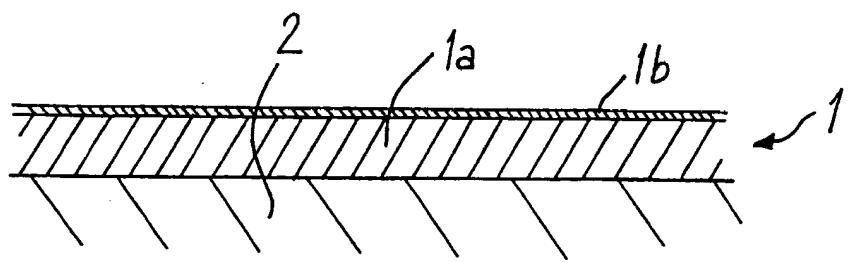
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10. A material according to claim 9, **characterized** in that the thermoplastic polyolefin elastomer is an ethylene- α -olefin copolymer rubber, such as EPR, EBR, or EPBR.

10 11. A material according to any of the claims 1 to 10, **characterized** in that it contains cross-linked structures.

15 12. A material according to any of the previous claims, **characterized** in that on its surface there is a coating layer (1b), preferably a transparent coating layer.

1/1



INTERNATIONAL SEARCH REPORT

International application No.
PCT/FI 98/00296

A. CLASSIFICATION OF SUBJECT MATTER

IPC6: C08L 23/00, C08L 23/16, C08K 5/01 // E04F 15/10, D06N 7/02
 According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)

IPC6: C08L, C08K, E04F, D06N

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

SE,DK,FI,NO classes as above

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)

WPI

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages.	Relevant to claim No.
X	US 4166057 A (HAYASHI TAKEMORI), 28 August 1979 (28.08.79), column 1, line 9 - line 18; column 8, line 16 - line 29; column 8, line 60 - line 61, column 11, line 49 - line 58 --	1-12
A	WO 9604419 A1 (FORBO-NAIRN LIMITED), 15 February 1996 (15.02.96), page 5, line 6 - line 32; page 6, line 15 - line 30; page 9, line 9 - line 25, page 15, line 28 - line 30; page 18, line 16 - line 22; page 22, line 28 - line 31; page 25, line 11 - line 27 --	1-12
A	DE 4436486 A1 (DLW AG), 18 April 1996 (18.04.96) --	1-12

Further documents are listed in the continuation of Box C.

See patent family annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
6 July 1998	07-07-1998
Name and mailing address of the ISA/ Swedish Patent Office Box 5055, S-102 42 STOCKHOLM Facsimile No. + 46 8 666 02 86	Authorized officer Monika Bohlin Telephone No. + 46 8 782 25 00

INTERNATIONAL SEARCH REPORT

International application No.
PCT/FI 98/00296

C (Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

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A	WO 9710298 A1 (TARKETT AB), 20 March 1997 (20.03.97), page 4, line 8 - line 16, abstract --	1-12
A	WO 9415014 A1 (UPOFLOOR OY), 7 July 1994 (07.07.94), abstract, claims --	1-12
A	EP 0300689 A2 (BP CHEMICALS LIMITED), 25 January 1989 (25.01.89), page 4, line 13 - line 42 -- -----	1

INTERNATIONAL SEARCH REPORT
Information on patent family members

30/06/98

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